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UNIVERSITY OF COLORADO
BOULDER, COLORADO 80309

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SEMIANNUAL REPORT

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Principal Investigators:
A.V. Phelps
Telephone: (303) 492-7850
A.C. Gallagher
Telephone: (303) 492-7841
Scientific Officer: Director,
Physics Program, ONR
Short Title of Work: Laser Processes

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Cable Address: JILA

Telephone Number: 492-7789

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The modeling of electrical discharges in high pressure Na-Xe mixtures for high power laser applications has been extended to include stimulated emission by incident radiation at the wavelength of the excimer band. In addition, we have explored the effects of the use of very high sodium densities and of changes in the products of dissociative recombination. It is concluded that an efficient laser amplifier would result if one could find an excimer mixture with favorable products of dissociative recombination.

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The experimental evaluation of metal vapor-rare gas mixtures has been extended to include Mg-Xe mixtures. We obtain stable discharges similar to those in Na-Xe mixtures. An apparent excimer band extends far to the red side of the resonance line of Mg at 285 nm. The large extent of this red wing suggests that it will be relatively easy to obtain gain using the Mg Xe mixture. It remains to be seen whether the gain will be large enough to be useful and as to whether there are interfering absorption bands.

Measurements of the rate coefficients for electron excitation of the $N_2(A^3\Sigma)$ metastable state in pure N_2 have been completed. The measured excitation rate coefficients are somewhat smaller than predicted using electron collision cross sections determined from electron transport data. In order to obtain agreement between calculations and experiment it is necessary to increase the cross sections for vibrational excitation of nitrogen by about a factor of 1.3 over values obtained from electron transport data.

SEMIANNUAL REPORT

This Semiannual Report contains descriptions of work carried out under ONR Contract No. N00014-76-0123 and ARPA Order No. 2683-Amd. 6. It covers the period from 1 February 1978 to 31 July 1978. Section I is the Semiannual Report Summary while Sections II-III are more detailed descriptions of work carried out under the projects supported by this contract.

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I. SEMIANNUAL REPORT SUMMARY

The two projects being carried out under this contract are summarized below. More detailed discussions are given in Sections II and III of this report.

(1) Metal Vapor-Rare Gas Discharges.

The objective of this project is the evaluation of the potential of electrical discharges in high pressure metal vapor-rare gas mixtures for excitation of high power, high efficiency lasers operating at near visible wavelengths. Our previous work has shown that many metal vapor-rare gas excimers have reasonable stimulated emission coefficients at visible or near visible wavelengths and that electrical discharges with sufficiently high electron temperatures (>0.5 eV) should be an efficient way to produce these excimers. Our recent experiments have shown that the effective electron temperature in high power discharges in Na-Xe mixtures with Na densities between 10^{15} and 10^{16} cm^{-3} is too low to yield gain on the Na-Xe excimer transition. Work carried out during this report period suggests that operation of these discharges at much higher Na densities, e.g., 3×10^{17} cm^{-3}, may allow operation of the discharge at significantly higher electron temperatures. These models continue to show the need for a large energy loss in the dissociative recombination process. The search for a metal vapor system with more desirable recombination properties than those of the alkalis has led us to experimental measurements in the Mg-Xe system. Our initial measurements show that discharges in Mg-Xe at atom and current densities similar to those used in the Na-Xe experiments are stable for several microseconds without the necessity for preionization. The presence of an extended red wing to the resonance line at 285 nm suggests that it will be relatively easy to obtain gain in the Mg-Xe system. We have

not yet determined the excitation temperature or the absorption coefficients for these discharges and so cannot predict their usefulness for high power laser applications.

(2) Electron Excitation of Metastable Atoms and Molecules.

The technical problem addressed in this project is the measurement and prediction of rate coefficients for the electron excitation of the $A^3\Sigma_u^+$ metastable state of N_2 and the metastable states of Ar. The metastable N_2 molecules have been suggested by DoD contractors as an efficient source of excitation for the upper laser levels of molecules and atoms, such as NO and Hg, while the Ar metastable states are one of the two important sources of excitation in most KrF and XeF lasers, e.g., those utilizing discharge enhancement in mixtures of Ar, Xe and F_2. Our measurement of the rate coefficients for the electron excitation of the $N_2(A^3\Sigma)$ metastable states of N_2 in pure nitrogen yield values somewhat smaller than predicted using electron collision cross sections determined from electron transport data. Agreement between calculation and experiment can be obtained by increasing the cross sections for vibrational excitation of nitrogen by about a factor of two over values which we had obtained a number of years ago, and by about a factor of 1.3 larger than values we have obtained in recent analyses of electron transport data. The source of the discrepancy between these two values is unknown.

II. METAL VAPOR-RARE GAS DISCHARGES

Drs. W. L. Morgan, R. Shuker (to 5/78), L. Shumann, A. Gallagher and A. V. Phelps.

During this report period our experimental project was directed toward the evaluation of the potential of the Mg-Xe system for nigh power visible laser applications. As we have indicated previously, the Mg-Xe excimer is expected to have good optical properties while the relatively high vapor pressure of Mg makes it possible to operate the discharges in the same experimental apparatus as used for Na-Xe. The Mg-Xe molecule is expected to be representative of excimers formed from metals of column IIa of the periodic table. Our modeling effort during this report period was concerned with the effect of the irradiation of discharges such as those in Na-Xe with high intensity radiation at wavelengths near those of maximum excimer gain. The Na-Xe models developed earlier were used for these calculations and are considered representative of what will happen with other discharge excited excimer systems.

We have measured the emission spectrum of a high power pulsed discharge in Mg doped Xe. As in previous studies of Na doped Xe we obtain a stable, steady-state discharge for several μs without the necessity of preionization or e-beam sustaining. Since most of the pulse energy is delivered during the steady-state portion of the discharge we are concentrating on the properties during this time. The emission spectrum for a Mg density [Mg] $\cong 1 \times 10^{16}$ cm^{-3}, xenon density [Xe] = 2.1×10^{19} cm^{-3}, and current density J = 175 Amp/cm^2 is shown in Fig. 1. We have not yet studied a full set of [Mg], [Xe], and J, but by lowering [Xe] by about a factor of 100 we have been able to identify the strong bands in the 500 and 540 nm regions as due to Mg_2. Also,

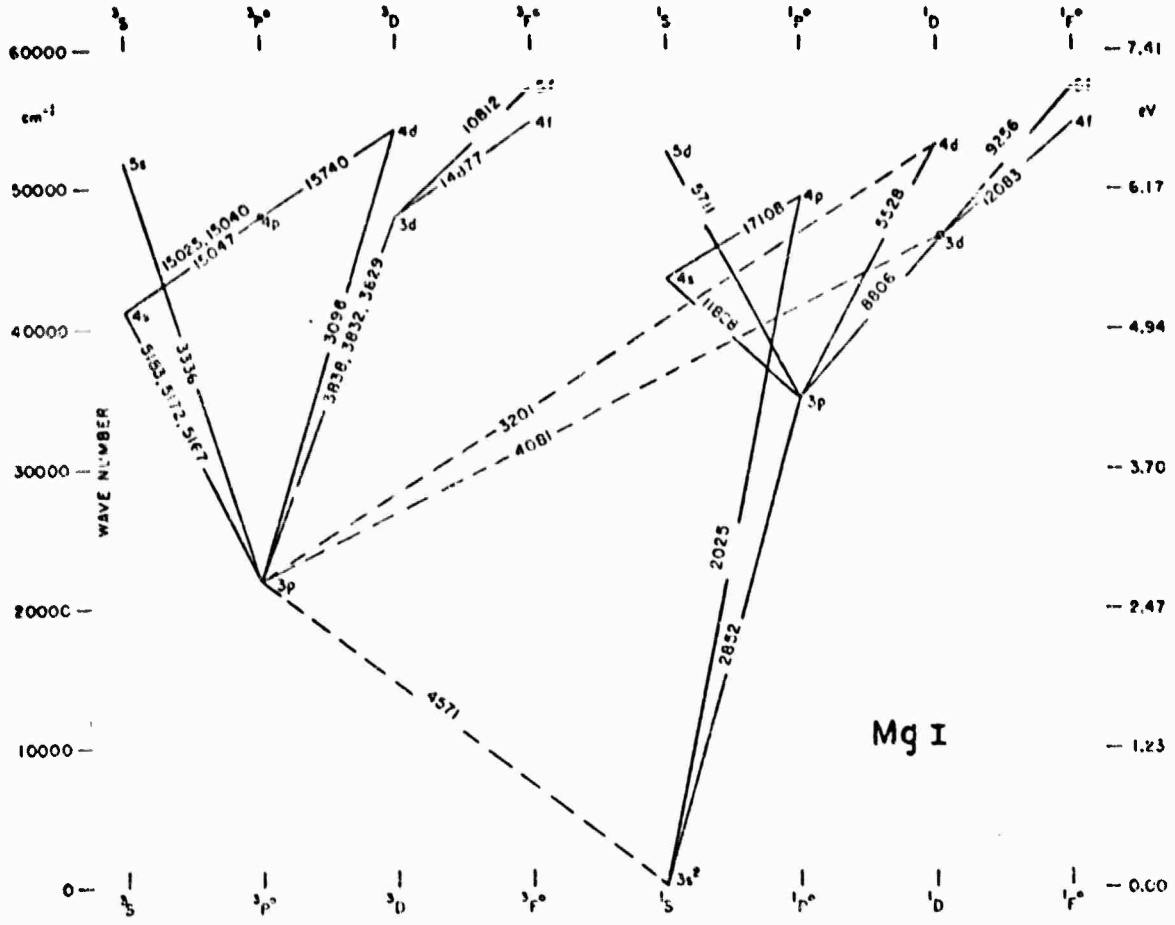

Figure 1. Energy levels for magnesium. The excimer band of potential laser interest is to the red of the 285.2 nm ($3^1P \rightarrow 3^1S$) resonance line.

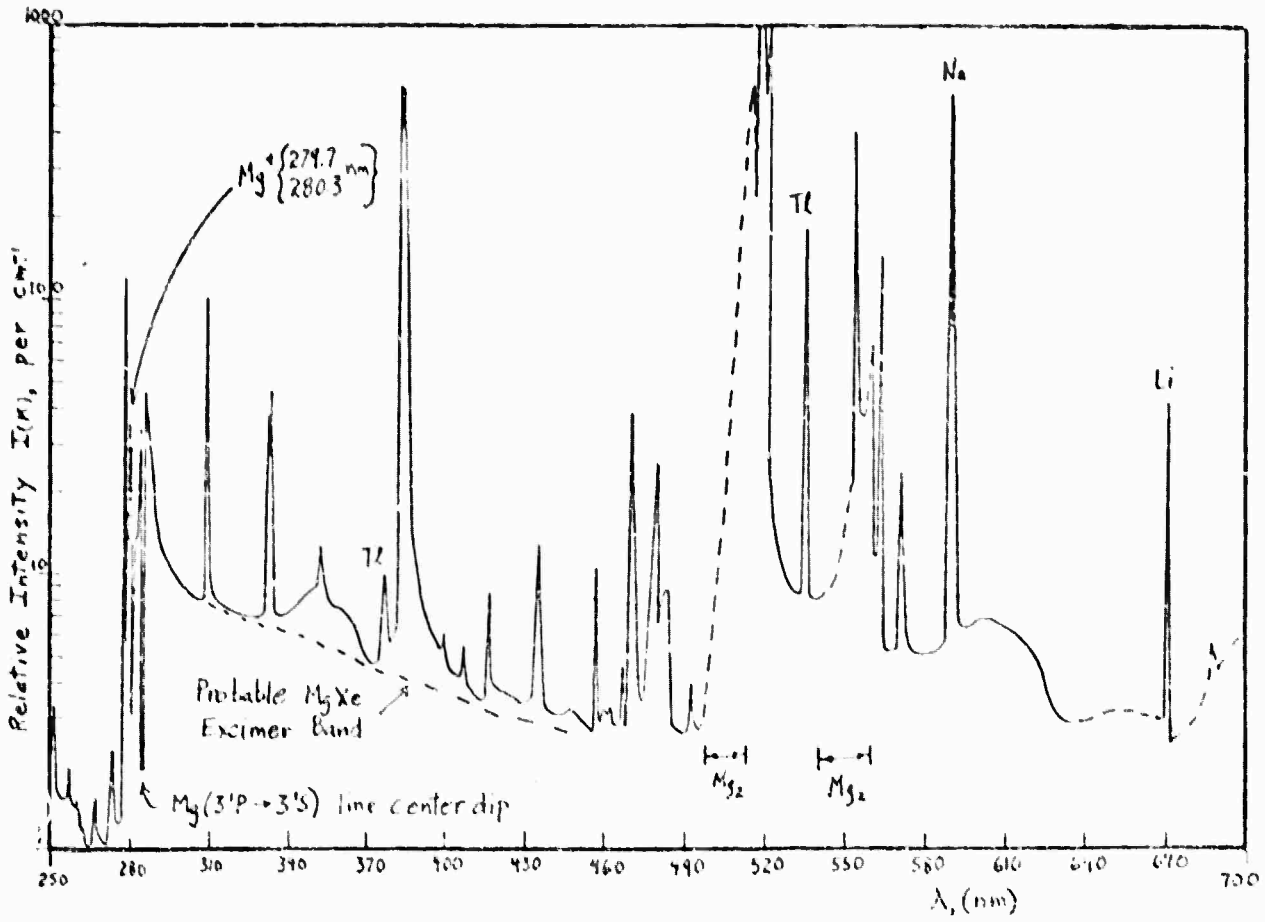

Figure 2. Relative spectral intensity observed from Mg-Xe discharge.

on the basis of our present, incomplete results we have identified the probable shape of the Mg-Xe excimer band associated with the resonance line at 285 nm. Figure 1 shows the 3^1P-3^1S transition in Mg as well as the important atomic transitions. As can be seen in Fig. 2 this band extends very far to the red of the resonance line. This is very favorable for providing useful gain in the long-wavelength portions of the band, where net gain on the excimer band is most easily achieved. However, more data is needed to reliably ascertain the excimer-band stimulated emission coefficient in the blue and blue-green regions. Lines due to minor impurities of Na, Li, and Tℓ are identified in Fig. 1; these constituents are deemed too minor to significantly influence the discharge. Lines of Mg^+ and Xe are also indicated; the remaining lines are due to transitions between excited states of Mg. The intensities of these lines can be used to determine excited state densities and thereby the electron temperature and excitation temperature in the discharge. The discharge shape appears much like the sodium-xenon discharge with a bright cathode spot spreading into a green homogeneous positive column. The main difference is that the Mg-Xe positive column is slightly narrower than for sodium under the same particle density and current density conditions. The E/N are not yet accurately determined but are of the same magnitude as for Na doped Xe.

The effects of high intensity radiation on the optical and electrical properties of high powered discharges in metal vapor-rare gas mixtures were investigated using the models developed earlier for the Na-Xe system. As shown by the comparison of theory and experiment cited in the last Semiannual Report our experiments seem to require that the products of the dissociative recombination of electrons and Na_2^+ ion be an atom in the ground state and an atom in a highly excited state, e.g., a state with effective

principal quantum number n near 6. Our simplified analytical models showed that if the excited atom were produced in a lower excited state, e.g., Na(3P), the discharge would have a much higher gain and electrical impedance. We have therefore investigated the laser properties of a still to be discovered, metal-vapor based discharge in which the metal is sodium-like except for dissociative recombination into a strongly bound state rather than the n=6 states. Although this commonly occurs in noble gas recombination, it remains to be determined whether a real metal vapor excimer with such properties can be found.

The results of our modeling of the modified Na-Xe system are shown in Figs. 3 and 4. The model has 19 excited states, 5 charged species, and approximately 400 rate coefficients. Figure 3 shows the ground state and excited state densities normalized to their respective statistical weights as a function of the excitation potential of the atomic levels. The electron temperature is chosen to be high enough, i.e., 0.52 eV, so that significant net gain is obtained. Figure 3 shows the change in the normalized excited state populations as the incident radiant flux is increased. Figure 3 shows that for input radiant fluxes at 700 nm and at intensity levels below 320 MW/cm^2 the excited state populations are rather close to the values predicted for Saha equilibrium at the electron temperature of .52 eV. Figure 4 shows that at these lower input fluxes the radiant power output increases approximately linearly with the input power, and that the efficiency of conversion of electrical energy into radiant energy is low. At input fluxes above about 330 MW/cm^2 there is a sudden change in the operating characteristics of the discharge until at 560 MW/cm^2 Fig. 3 shows that the density of the higher excited states, and therefore of the electrons, drops well below the Saha equilibrium value. Figure 4 shows that for the higher input fluxes the power output decreases somewhat but that the efficiency of conversion of electrical input energy to radiant energy increases to 70%. Note the high Na densities used here.

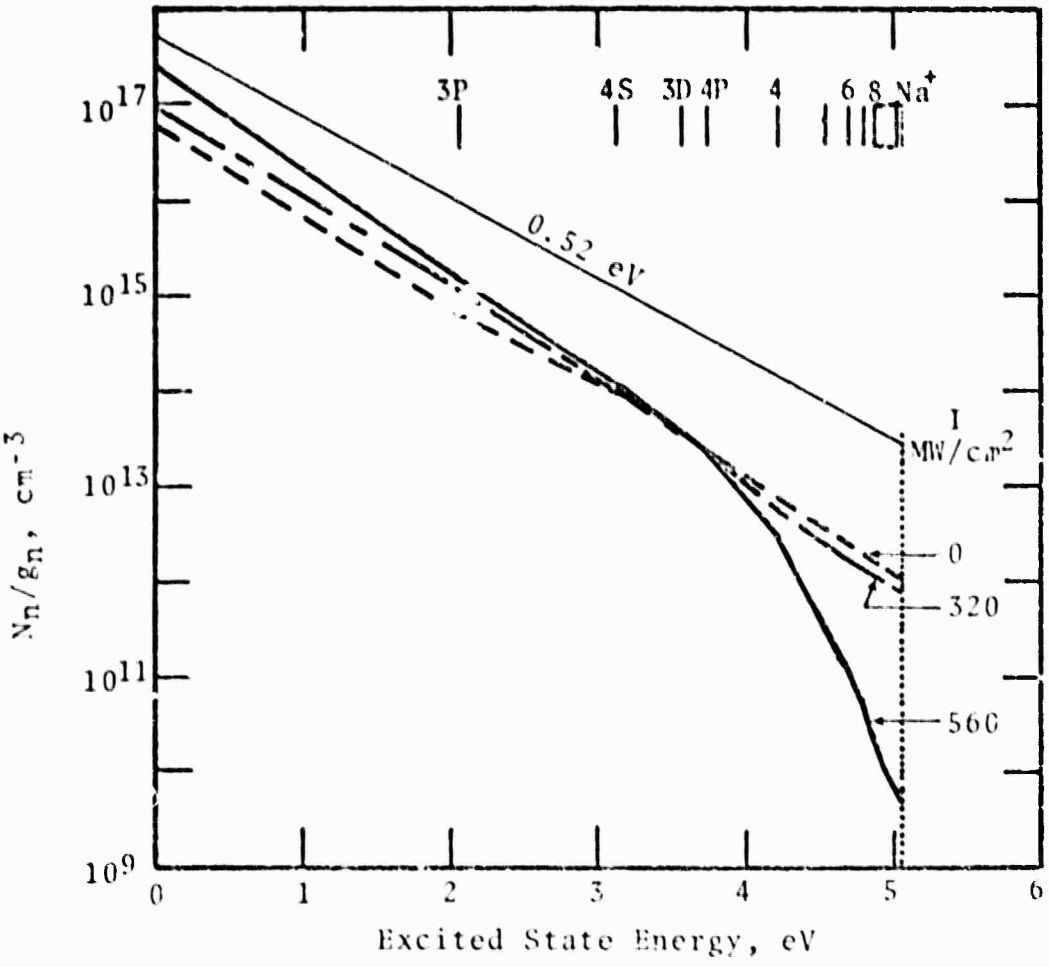

Figure 3. Calculated normalized excited state density vs. excited state energy for various incident radiant fluxes. Model uses Na-Xe coefficients except for dissociative recombination. See text. $[Na]_o = 2.7 \times 10^{17}$ cm^{-3}, $[Xe] = 2.7 \times 10^{20}$ cm^{-3} and $T_e = 0.52$ eV. The thin line labeled 0.52 eV shows the relative (not absolute) normalized populations expected for Saha equilibrium at the electron temperature. The vertical bars at the top of the figure show the energies of the lower excited states used in the model. The vertical dotted line is at the ionization potential of Na.

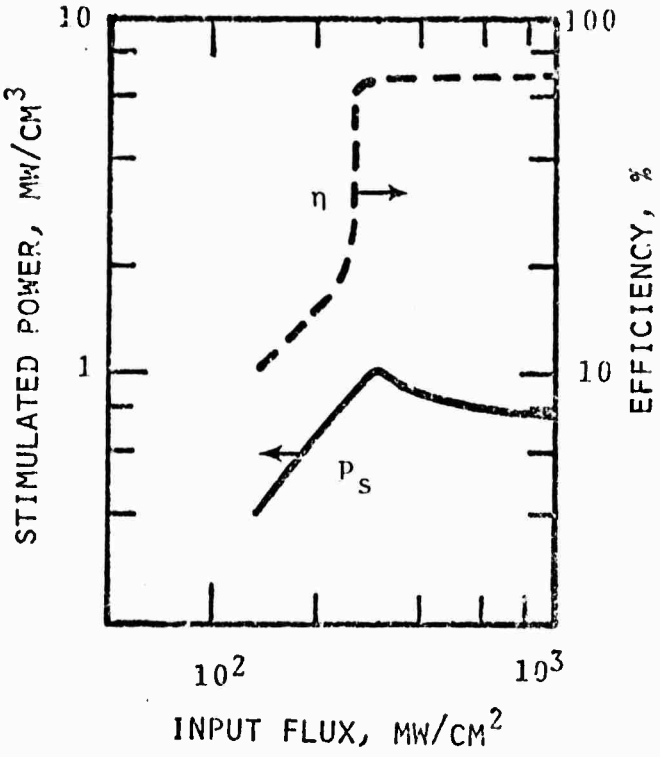

Figure 4. Calculated stimulated power output and efficiency for laser amplifier using modified Na-Xe model. See text, p. 9.

Our model shows that this drastic change in discharge occurs when the rate of destruction of Na(3P) atoms and of Na-Xe excimers by stimulated emission equals the rate of deexcitation by electron impact. The resultant decrease in the density of Na(3P) atoms is responsible for the two-order of magnitude decrease in the electron density. Note that not only is the efficiency high at these lar nput fluxes but that the discharge impedence is higher, making it possible to match the discharge to available power supplies much more efficiently. Somewhat similar effects occur when calculations are made for lower sodium and xenon densities, although in these cases the gain of the laser amplifier is much lower. Note also that this sudden improvement in discharge behavior is not indicated by the models when the products of dissociative recombination are sodium atoms in a highly state and in the ground state.

The preceding calculations support once again our assertion as to the importance of processes in which electrons recombine with molecular ions to produce strongly bound excited states. Dissociative recombination of electrons and molecular ions and the very similar process of predissociation of highly excited molecules are processes which have this potential. Other molecular deexcitation processes are under investigation. We believe that the potential of the metal based excimers for high efficiency, high power operation justify a continuous, vigorous search for systems with the desired electron-ion recombination properties along with good stimulated emission coefficients in the near visible region.

III. ELECTRON EXCITATION OF METASTABLE ATOMS AND MOLECULES

D. Levron and A. V. Phelps.

During this report period measurements of electron excitation rate coefficients for the $N_2(A^3\Sigma_u^+)$ metastable state have been completed. These measurements were made by observing the absolute intensity of radiation emitted by the metastable molecule in the Vegard-Kaplan band. Depending upon experimental conditions radiation was observed from the v=0 and v=1 vibrational states of the molecule. Using the kinetic data discussed in the last Semiannual Report, we have shown that approximately half of the metastable molecules are produced in the v=0 level and about half in the v=1 level. We have not been able to observe the cascading from higher vibrational levels and higher electronic states which leads to the production of these low-lying vibrational levels. The sum of the electron excitation rate coefficients for all processes leading to the production of the metastable state is shown as a function of the variable in N/E in Fig. 5. We have chosen this method of plotting the data because it spreads out the results and has an approximate straight line dependence of the logrithim of the excitation coefficient as a function of N/E. The solid points show the experimental results and the smooth curves show the predictions made using various sets of electron collision cross sections for nitrogen. The upper curve shows predictions made using the cross sections of Engelhart, Phelps, and Risk[1] as modified by deletion of their so-called 5.0 eV energy loss process. The middle curve shows the results of calculations in which the set of vibrational excitation cross sections used by Engelhart, Phelps and Risk has been multiplied by a factor of 1.5 for electron energies above 1.7 eV. This multiplicative factor was chosen so as to yield agreement between calculated and measured electron transport coefficients for nitrogen. Thus, the

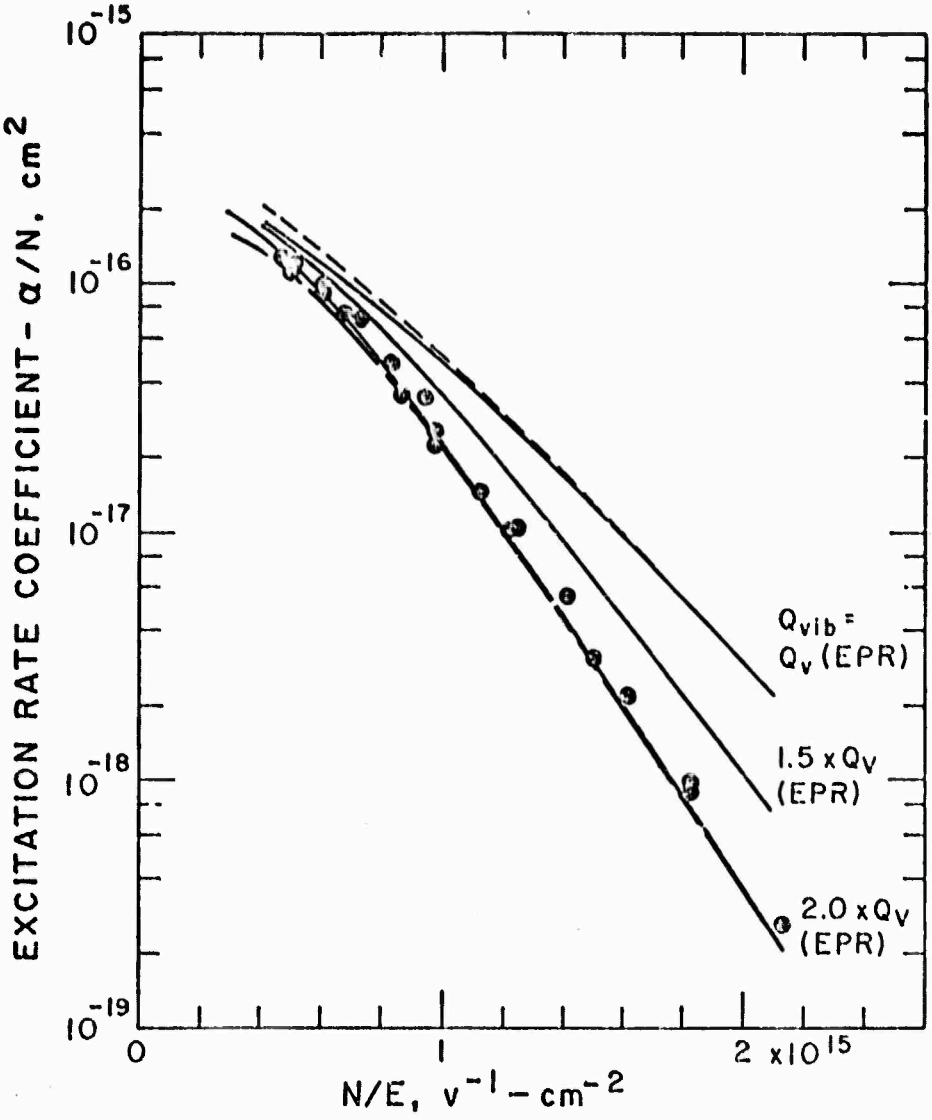

Figure 5. Electron excitation rate coefficients for $N_2(A^3\Sigma_u^+)$ metastables in pure N_2.

increased vibrational excitation cross section has compensated for the deletion of the spurious 5.0 eV energy loss used in the earlier calculation. From Fig. 5 we see that the use of the factor of 1.5 leads to a very significant over-estimate of the excitation rate coefficients for the $N_2(A\Sigma)$ metastable state. The lower curve shows the calculated rate coefficients when the multiplicative factor is increased to 2.0. The use of this factor leads to a slight underestimate of the rate coefficient for metastable excitation, so that a factor of about 1.9 should give a good fit to the experimental data at values of N/E above about 8×10^{14} V^{-1}-cm^{-2}.

An additional result brought out by the comparison in Fig. 5 is that the rate coefficient for electron excitation of the N_2 metastable state is very insensitive to the cross section for electronic excitation used in the model. Thus, the lower dashed curve shows the effect of reducing the electronic excitation cross sections of Cartwright et al. by a factor of 0.6. The upper dashed curve shows the effect of increasing these electronic excitation cross sections by a factor of 1.7. These comparisons therefore demonstrate that for N/E values of greater than about 4×10^{14} V^{-1}-cm^{-2}, or E/N values less than about 2.5×10^{-15} V-cm^2, the rate of excitation of the nitrogen metastables depends primarily on the ability of the electrons to gain enough energy to escape vibrational excitation at energies near 2 eV.

During the next report period we will begin measurements of metastable excitation coefficients in N_2-Ar. In these measurements we will attempt to measure electron excitation rate coefficients for argon metastables by measuring the rate coefficients for excitation of the nitrogen metastables in the N_2-Ar mixtures.

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Reference:

1. A. G. Engelhardt, A. V. Phelps and C. G. Risk, Phys. Rev. 135, A1566 (1964).